Supplementary Information

Scanning DMA Data Analysis

II. Integrated DMA-CPC Instrument Response and Data Inversion

Huajun Mai[*], Weimeng Kong[†], John H. Seinfeld[*,†], Richard C. Flagan[*,†,‡]

1 Integrated DMA-CPC instrument response

The total number of particles recorded in the SEMS in time bin i is

$$\begin{aligned} R_{i,\mathrm{SEMS}} &= Q_{\mathrm{a}} \int_{(i-1)t_{\mathrm{c}}}^{it_{\mathrm{c}}} \int_{-\infty}^{\infty} n(u) \sum_{\phi} p_{\mathrm{charge}}(u,\phi) \eta_{\mathrm{F}}(u,\phi) \eta_{\mathrm{CPC}}(u,\phi) \\ &\times \left[\Gamma_{\mathrm{SEMS}}(Z_{\mathrm{p}}(u,\phi),\beta,\delta,it_{\mathrm{c}}) - \Gamma_{\mathrm{SEMS}}(Z_{\mathrm{p}}(u,\phi),\beta,\delta,(i-1)t_{\mathrm{c}})\right] \mathrm{d}u, \end{aligned} \tag{1}$$

where Q_{a} is the incoming aerosol sample flow rate, and $n(u)$ is the size distribution of the source particles in terms of the logarithm of the particle diameter, $u = \log D_{\mathrm{p}}$. $p_{\mathrm{charge}}(u,\phi)$,

[*]Division of Engineering and Applied Science, California Institute of Technology, Pasadena, California 91125, USA

[†]Division of Chemistry and Chemical Engineering, California Institute of Technology, Pasadena, California 91125,USA

[‡]Corresponding author: flagan@caltech.edu

$\eta_{\rm F}(u,\phi)$, and $\eta_{\rm CPC}(u,\phi)$ are the probability that a particle of size u will acquire ϕ charges in the charge conditioner, the particle penetration efficiency, and the CPC counting efficiency, respectively. For convenience, we express the overall detection efficiency for particles of diameter u and ϕ charges concisely as $\eta(u,\phi) = p_{\rm charge}(u,\phi)\eta_{\rm F}(u,\phi)\eta_{\rm CPC}(u,\phi)$. $\Gamma_{\rm SEMS}(Z_{\rm p}(u,\phi),\beta,\delta,t)$ is the cumulative transfer function for the SEMS.

With the definition of the kernel function

$$h_i(u) \;=\; Q_{\rm a}\sum_{\phi}\eta(u,\phi)\left[\Gamma_{\rm SEMS}(Z_{\rm p}(u,\phi),\beta,\delta,it_{\rm c})-\Gamma_{\rm SEMS}(Z_{\rm p}(u,\phi),\beta,\delta,(i-1)t_{\rm c})\right], \quad (2)$$

the instrument response in Eq. (1) becomes

$$R_{i,\rm SEMS}=\int_{-\infty}^{\infty}h_i(u)n(u){\rm d}u. \quad (3)$$

The inversion problem is to retrieve the size distribution function $n(u)$ on the targeted particle size node $u_j^\dagger=\log D_{{\rm p},j}^\dagger$ $(j=1,2,\cdots,J)$. For purpose of data inversion, the particle size distribution is approximated using linear splines, which are described on the size interval $[u_j^\dagger,u_{j+1}^\dagger)$, as

$$\begin{aligned} n_j(u) &= n(u_j^\dagger)+\frac{n(u_{j+1}^\dagger)-n(u_j^\dagger)}{u_{j+1}^\dagger-u_j^\dagger}\left(u-u_j^\dagger\right) \\ &= n(u_j^\dagger)\frac{u_{j+1}^\dagger-u}{u_{j+1}^\dagger-u_j^\dagger}+n(u_{j+1}^\dagger)\frac{u-u_j^\dagger}{u_{j+1}^\dagger-u_j^\dagger}. \end{aligned} \quad (4)$$

The Fredholm integral, Eq.(3), thus becomes

$$R_{i,\rm SEMS}=\sum_{j=1}^{J-1}\int_{u_j}^{u_{j+1}}h_i(u)n_j(u){\rm d}u, \quad (5)$$

which is numerically evaluated by applying the trapezoidal rule on a particle-size grid $u_k=\log D_{{\rm p},k}$ $(k=1,2,\cdots,K)$. To improve the accuracy of the kernel calculation, the integration grid is much finer than the targeted size node, *i.e.*, $K\gg J$. In practice, the particle size grid u_k is created by slicing the targeted particle size interval $[u_j^\dagger, u_{j+1}^\dagger)$ into smaller size bins. Using trapezoidal integration, Eq.(3) can be written as a summation,

$$R_{i,\,\mathrm{SEMS}} = \sum_{k=1}^{K} \Delta u_k h_i(u_k) n(u_k), \tag{6}$$

where

$$\Delta u_k = \begin{cases} \dfrac{u_2 - u_1}{2}, & k = 1 \\ \dfrac{u_{k+1} - u_{k-1}}{2}, & k = 2, 3, \cdots, K-1 \\ \dfrac{u_K - u_{K-1}}{2}, & k = K \end{cases} \tag{7}$$

is the weighting factor arising from the trapezoidal integral. Combining Eqs.(4) and (6), the instrument response becomes

$$\begin{aligned} R_{i,\,\mathrm{SEMS}} &= \sum_{k=1}^{K} \Delta u_k \left[n(u_j^\dagger) \frac{u_{j+1}^\dagger - u_k}{u_{j+1}^\dagger - u_j^\dagger} + n(u_{j+1}^\dagger) \frac{u_k - u_j^\dagger}{u_{j+1}^\dagger - u_j^\dagger} \right] h_i(u_k), \\ &\qquad u_k \in [u_j^\dagger, u_{j+1}^\dagger). \end{aligned} \tag{8}$$

For each u_k, Eq. (8), we must first determine to which size interval $[u_j^\dagger, u_{j+1}^\dagger)$ it belongs, and then calculate the $h_i(u_k)$ values, and perform the summation. This process can be viewed from another perspective, in which we focus on the targeted size interval $[u_j^\dagger, u_{j+1}^\dagger)$ and then we find all u_k within the j^{th} interval, *i.e.*, $u_k \in [u_j^\dagger, u_{j+1}^\dagger)$. The summation index is then changed from k to j, leading to

$$\begin{aligned} R_{i,\,\mathrm{SEMS}} &= \sum_{j=1}^{J} \left[\sum_{u_k \geq u_j^\dagger}^{u_k < u_{j+1}^\dagger} \Delta u_k \frac{u_k - u_j^\dagger}{u_{j+1}^\dagger - u_j^\dagger} n(u_{j+1}^\dagger) h_i(u_k) + \sum_{u_k \geq u_j^\dagger}^{u_k < u_{j+1}^\dagger} \Delta u_k \frac{u_{j+1}^\dagger - u_k}{u_{j+1}^\dagger - u_j^\dagger} n(u_j^\dagger) h_i(u_k) \right] \\ &= \sum_{j=1}^{J} \left[\sum_{u_k \geq u_{j-1}^\dagger}^{u_k < u_j^\dagger} \Delta u_k \frac{u_k - u_{j-1}^\dagger}{u_j^\dagger - u_{j-1}^\dagger} n(u_j^\dagger) h_i(u_k) + \sum_{u_k \geq u_j^\dagger}^{u_k < u_{j+1}^\dagger} \Delta u_k \frac{u_{j+1}^\dagger - u_k}{u_{j+1}^\dagger - u_j^\dagger} n(u_j^\dagger) h_i(u_k) \right] \\ &= \sum_{j=1}^{J} \left[\sum_{u_k \geq u_{j-1}^\dagger}^{u_k < u_j^\dagger} \Delta u_k \frac{u_k - u_{j-1}^\dagger}{u_j^\dagger - u_{j-1}^\dagger} h_i(u_k) + \sum_{u_k \geq u_j^\dagger}^{u_k < u_{j+1}^\dagger} \Delta u_k \frac{u_{j+1}^\dagger - u_k}{u_{j+1}^\dagger - u_j^\dagger} h_i(u_k) \right] n(u_j^\dagger), \end{aligned} \tag{9}$$

This instrument response equation can be expressed in matrix form as $\mathbf{R} = \mathbf{A}\mathbf{N}$, where $\mathbf{R} = [R_{1,\mathrm{SEMS}}, R_{2,\mathrm{SEMS}}, \cdots, R_{I,\mathrm{SEMS}}]^{\mathrm{T}}$ is the instrument response time-series , $\mathbf{N} = [n(u_1^\dagger), n(u_2^\dagger), \cdots, n(u_J^\dagger)]^{\mathrm{T}}$ is the vector of values representing the size distribution, and $\mathbf{A}$ is the kernel matrix, with elements

$$\mathbf{A}_{i,j} = \sum_{u_k \geq u_{j-1}^\dagger}^{u_k < u_j^\dagger} \Delta u_k \frac{u_k - u_{j-1}^\dagger}{u_j^\dagger - u_{j-1}^\dagger} h_i(u_k) + \sum_{u_k \geq u_j^\dagger}^{u_k < u_{j+1}^\dagger} \Delta u_k \frac{u_{j+1}^\dagger - u_k}{u_{j+1}^\dagger - u_j^\dagger} h_i(u_k). \tag{10}$$

2 Integrated SEMS system response plots

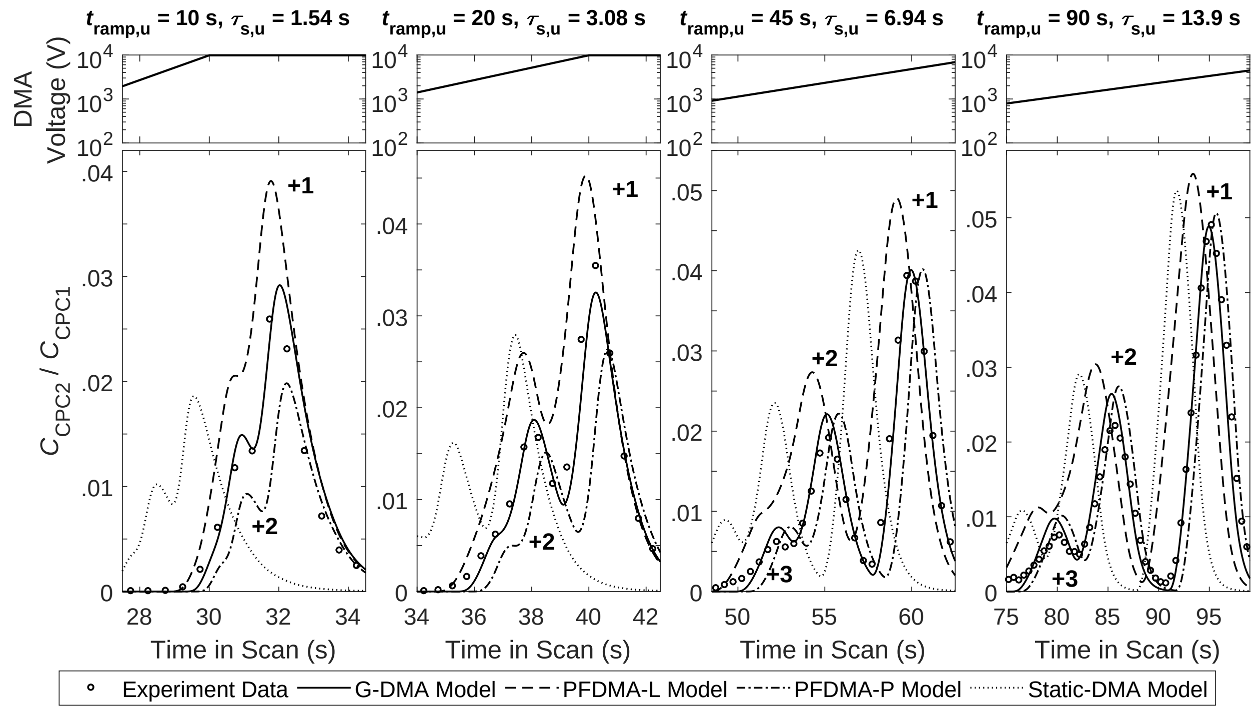

Figure S1: Up-scan experimental and modeling results for SEMS instrument response to monodisperse 296 nm particles with ramp duration $t_{\mathrm{ramp}} = 10$, 20, 45 and 90 s (corresponding to scan time $\tau_{\mathrm{s}} = 1.54$, 3.08, 6.94 and 13.9 s).

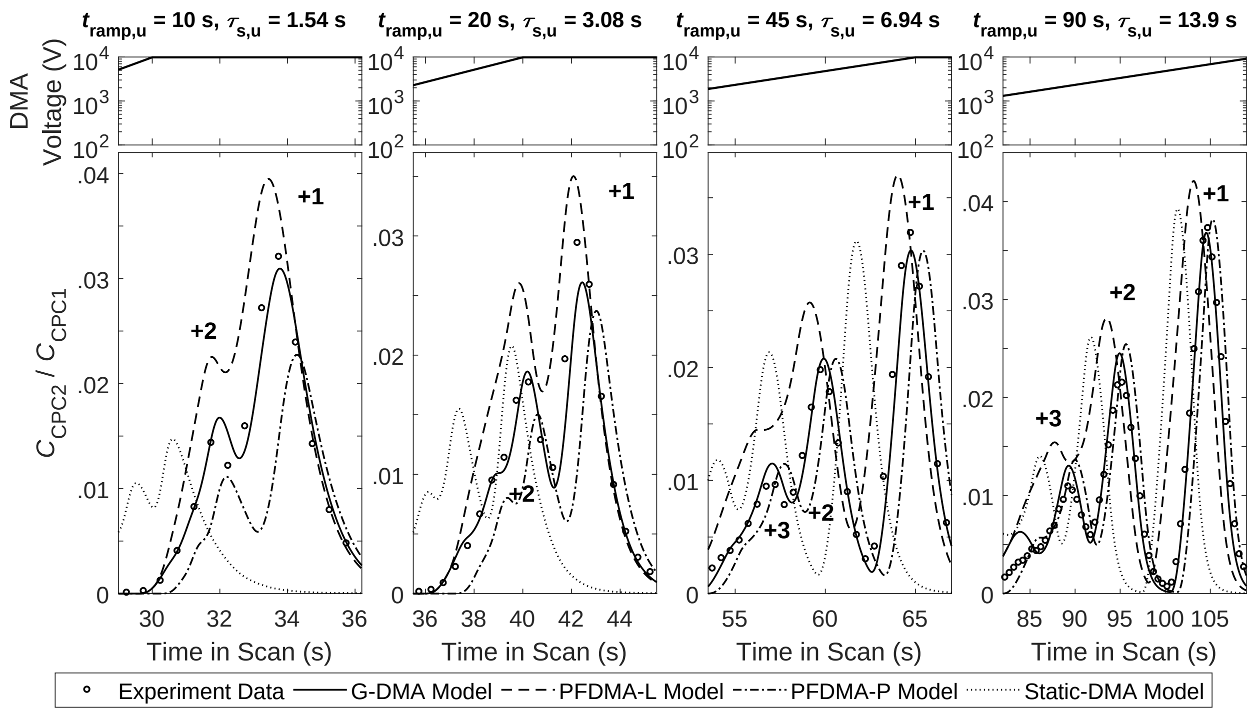

Figure S2: Up-scan experimental and modeling results for SEMS instrument response to monodisperse 498 nm particles with ramp duration $t_{\mathrm{ramp}} = 10$, 20, 45 and 90 s (corresponding to scan time τ_{s} = 1.54, 3.08, 6.94 and 13.9 s).

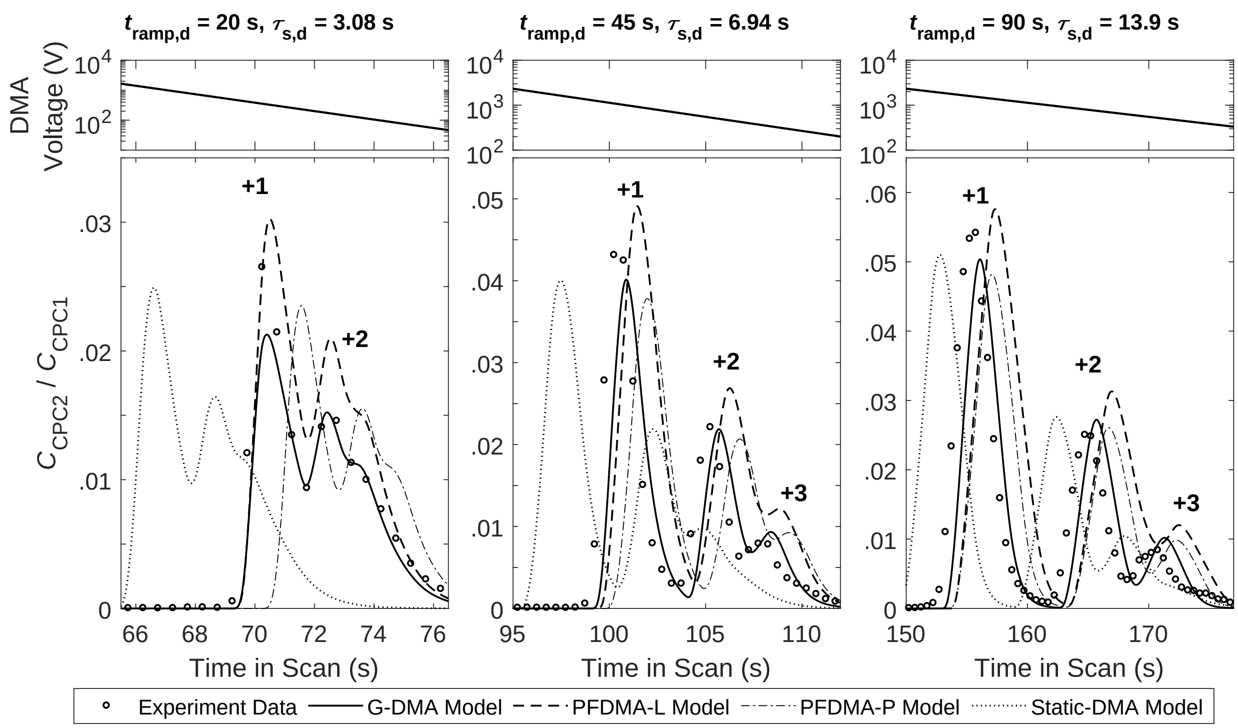

Figure S3: Down-scan experimental and modeling results for SEMS instrument response to monodisperse 296 nm particles with ramp duration $t_{\mathrm{ramp}} = 10$, 20, 45 and 90 s (corresponding to scan time $\tau_{\mathrm{s}} = 1.54$, 3.08, 6.94 and 13.9 s).

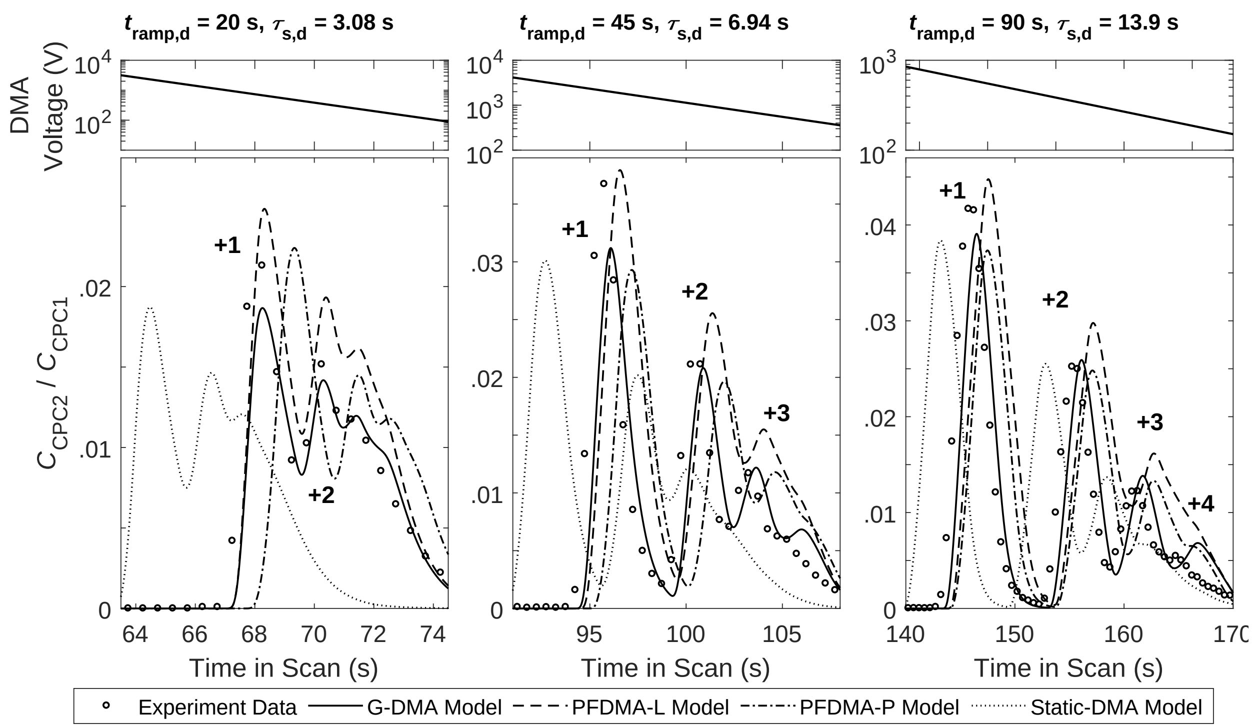

Figure S4: Down-scan experimental and modeling results for SEMS instrument response to monodisperse 498 nm particles with ramp duration $t_{\mathrm{ramp}} = 10$, 20, 45 and 90 s (corresponding to scan time $\tau_{\mathrm{s}} = 1.54$, 3.08, 6.94 and 13.9 s).